biased high because of additional neutrons from (α,n) reactions. Examples of (α,n)-reaction effects are given in Section 15.5.1. Active neutron techniques are not subject to the effects of passive backgrounds if the interrogation source is strong enough. However, active techniques can be strongly biased either high or low depending on the effects of matrix moderation and absorption on the incoming and outgoing neutrons. For a given waste-screening application, the choice of measurement technique should be made on the basis of cost, simplicity, sensitivity, and penetrability.

20.4 CONFIRMATORY MEASUREMENTS

20.4.1 Purpose

The transfer and storage of unirradiated nuclear materials is a frequent and large-scale activity at many NRC and DOE facilities. Many safeguards issues arise during the process of shipping, receiving, and inventory verification. Measurements can help to confirm that (a) material has not been diverted in transit, (b) the item identification is correct, (c) there is no undue radiation hazard to workers, and (d) inventory records are credible. Such confirmatory measurements may be simpler than measurements made for accountability purposes. For example, they require less time and less unpacking or repackaging of material. They also may be more versatile. However, in general, they are less accurate. Confirmatory measurements determine such attributes as weight, gamma-ray spectrum, total neutron radiation, and enrichment that—taken as a whole—are very difficult to imitate.

When nuclear material is transferred from one facility to another, present regulations require that the receiver verify the piece count, identification, and gross weight of the items in the shipment. Normally the receiver should perform accountability measurements on the items within 10 calendar days. In practice, however, this is often difficult to achieve because of (a) limitations in the availability of personnel and nondestructive assay equipment, (b) the length of time required for performing chemical analysis and transferring shipments into and out of storage vaults, and (c) the radiation exposure to personnel during packing and unpacking. Also, difficulty in measuring a relatively small number of scrap materials can delay closing the material balance on the shipment. One safeguards approach to alleviating these problems is to make confirmatory measurements at both the shipping facility and the receiving facility with similar or identical instruments (Ref. 7). Such measurements can confirm that there are no missing, incorrect, or bogus items in the shipment.

When nuclear material is stored at a facility, present regulations require periodic inventory of the entire facility and its storage vault. Confirmatory measurements made during that time on a random sample of the inventory can help identify mislabeled items and increase the credibility of the inventory process (Ref. 8).

20.4.2 Nondestructive Assay Options

Nondestructive assay techniques are well suited for confirmatory measurements because of their speed and their ability to measure an entire item. In some cases, it is also

possible to measure the shipping container itself, although with some loss of precision and accuracy. Options based on nondestructive assay of plutonium or uranium radiation attributes are summarized in this section.

For most plutonium samples, a combination of calorimetry and gamma-ray spectroscopy provides the best available accuracy: 0.5 to 2% for homogeneous materials. However, this instrumentation is usually reserved for accountability measurements because of its relatively high cost, complexity, and low throughput. Passive gamma-ray counting of the 414-keV ^{239}Pu peak in either a far-field geometry or by segmented scanning is a simpler option for materials of low density. However, most plutonium-bearing materials that are attractive for diversion are too dense for gamma-ray counting and are best measured by passive neutron counting. The technique is relatively simple, and can sometimes be applied to 30- or 55-gal. shipping drums without unpacking their contents. The neutron well counter should have uniform efficiency over the volume of the sample. Also, the electronics deadtime should be small and well known so that count ratios can be determined accurately.

Confirmatory neutron measurements of plutonium can be based on total or coincident counting, but coincident counting is a more specific attribute. Counting times are in the range of 100 to 1000 s. Typical accuracies for quick confirmatory measurements are 1 to 10% for well-characterized materials, 25% for impure scrap, and 50% for heterogeneous materials with high (α,n) rates (Ref. 8). However, the repeatability of raw measurement results is approximately 1%. It would be very difficult technically to construct a bogus item with the same weight, total neutron count rate, and coincident neutron count rate as a real item. This is also true for heterogeneous materials with high alpha decay rates where the assay accuracy is poor but the neutron attribute measurement is quite precise.

For passive neutron measurements of plutonium, the following guidelines show how the observed count rates are related to specific material attributes:

(1) The total neutron count rate is proportional to fertile content but also depends on the (α,n) reaction rate. If the fertile content can be determined from the coincident count rate, then any "excess" total count rate can be attributed to chemical compounds or impurities.

(2) The neutron coincidence rate is proportional to fertile content, but may be enhanced by induced fissions.

(3) The coincidence/totals ratio is a function of sample self-multiplication and, indirectly, fissile content. For heterogeneous plutonium scrap with very strong (α,n) reactions, the coincidence/totals ratio may provide the best possible measure of ^{239}Pu content, perhaps within 10%, if an iterative correction for ^{240}Pu content is made (Ref. 9).

(4) The difference in coincident neutron response with and without a cadmium liner in the well counter, divided by total neutron response, is a measure of fissile content (Section 17.3.3 and Ref. 10).

Confirmatory measurements of uranium are more difficult than those of plutonium. The alpha-particle emission rates are not high enough to permit heat production measurements. Enrichment measurements are possible with the 186-keV gamma ray, but they sample only the surface of the material and require a well-collimated geometry outside of the shipping drum. Far-field gamma-ray measurements can be used for low-density materials. They have also been used to confirm high-density materials to within a factor of 2 (Ref. 8).

The measurement of bulk uranium samples requires the use of active neutron systems, with the simplest being the Active Well Coincidence Counter (AWCC) (Section 17.3.1). In the thermal mode the AWCC is appropriate for samples containing from 5 to 100 g of ^{235}U. In the fast mode the instrument is limited to samples containing 50 g of ^{235}U or more, even for 1000-s counting times, because of the high accidental coincidence background of the interrogation sources. Good coupling must be maintained between the sources and the uranium, which usually requires the use of small containers. Thus, active coincidence counting of uranium is not as versatile or as easy to apply as passive coincidence counting of plutonium.

Two specific applications of active neutron counting of uranium are summarized below:

(1) Mixed uranium/plutonium samples: The passive coincidence response is proportional to ^{240}Pu but may be enhanced by induced fission in ^{235}U. Correction for self-multiplication can compensate for induced fission but will not provide a direct measure of ^{235}U content. Determination of ^{235}U or ^{239}Pu fissile content is not practical by active coincidence counting and requires more complex active neutron systems.

(2) Highly enriched uranium in UF_6 cylinders: The coincidence/totals ratio is proportional to ^{235}U content to within 2 to 10% (Section 17.3.4).

20.4.3 Recent Experience

Several examples follow of recent confirmatory measurements at Hanford, Rocky Flats, Los Alamos, and Savannah River. The examples illustrate different approaches and different levels of accuracy; they are arranged roughly in order of increasing degrees of confirmation.

Verification of a wide variety of stored nuclear material has been obtained by performing confirmatory measurements on a random sample of the inventory (Ref. 8). Passive neutron coincidence counting of plutonium and passive gamma-ray counting of uranium in a far-field geometry were the preferred techniques. Roughly 5% of the measurements were invalidated because of poor counting statistics, unsuitable material matrices or geometries, or lack of appropriate standards. Another 5% were judged as not confirmed because of results inconsistent with those obtained earlier on similar items. For the latter 5%, a superior instrument or technique was used to perform an accountability measurement. In about half of these cases the more accurate accountability measurement verified that the original item label was indeed incorrect.

Confirmation of incoming plutonium scrap metal has been accomplished by passive neutron coincidence counting of "bird cage" shipping containers (Ref. 11). Measurement of the shipping container itself rather than the individual interior items resulted in an eightfold reduction in work hours and a thirtyfold reduction in radiation exposure. Measurement accuracy was roughly 5% (1σ) for the shipping container as a whole compared to 2.5% (1σ) for the individual items. The receiver was able to verify the incoming shipment within three working days.

Confirmatory measurements of plutonium oxide have been performed by both the shipper and the receiver, each using a neutron coincidence counter of different design (Ref. 8). The counters measured the individual cans outside of their shipping drums. No attempt was made to normalize the response of one counter to the other. Instead, the

confirmation was based on the ratio of the responses. The total neutron count ratios were consistent to 0.5% (1σ), and the coincidence count ratios were consistent to 1.5% (1σ) before and after shipment. The receiver also compared his measurement of the actual plutonium mass as obtained by coincidence counting with that obtained by calorimetry for eight batches of cans. This comparison was not as accurate, having a 4.1% (1σ) scatter. The reduced accuracy of the mass determination is attributed to differences in settling, oxide density, moisture, or isotopics between batches. For example, the coincidence response of a 1-kg plutonium oxide can will change by about 1% for a 5% change in density (see Figure 16.14 in Chapter 16).

Shipper and receiver confirmatory measurements of plutonium-bearing ash, sand, slag, crucible, and oxide have been carried out by segmented gamma scanners of different design (Ref. 8). Standards were fabricated by the shipper, calibrated on the shipper's calorimeter, and sent to the receiver. The receiver's measurements of ^{239}Pu content agreed with the shipper's measurements to within 1 to 4% (1σ).

Confirmatory measurements of impure plutonium metal and oxide have been made with two identical neutron coincidence counters that measure 30-gal.-drum shipping containers. Figure 20.1 shows cutaway views of one of the counters. The counters are the first instruments designed specifically for confirmatory measurements (Ref. 12). The design features two doors, drum rollers, a drum positioner, and void spaces in the polyethylene wall to flatten the vertical efficiency profile. Normalization of response between shipper and receiver is accomplished by exchange of ^{252}Cf sources, source measurement data, and background measurement data. The confirmatory measurements consist of three 100-s total neutron counts. Initial results provided a shipper/receiver verification within 2 to 3% for oxide and within 1% for metal (Ref. 11). There is some evidence of a small bias that may be due to settling of the contents during shipment.

20.5 NUCLEAR MATERIAL HOLDUP

The term "holdup" refers to the accumulation of nuclear material inside the processing equipment of nuclear facilities. Other common terms for such material are "hidden inventory," "normal operating loss," and "in-process inventory." The choice of terminology depends in part on the application or point of view. For example, the nuclear material that remains in the facility after the runout of all bulk product may be called "in-process inventory." The material that remains after thorough brushing, wiping, acid leaching, and rinsing may be called "fixed holdup."

Because of the high economic value of nuclear material and the need to ensure radiation safety and criticality safety and to safeguard against theft or diversion, it is important to minimize holdup, to measure or model its magnitudes, and to remove it. Holdup causes and mechanisms, holdup magnitudes, and holdup modeling and measurement techniques are discussed in the remaining sections of this chapter.

20.5.1 Causes and Mechanisms

Nuclear material tends to accumulate in cracks, pores, and regions of poor circulation within process equipment. In addition, the internal surfaces of pipes, tanks, ducts,

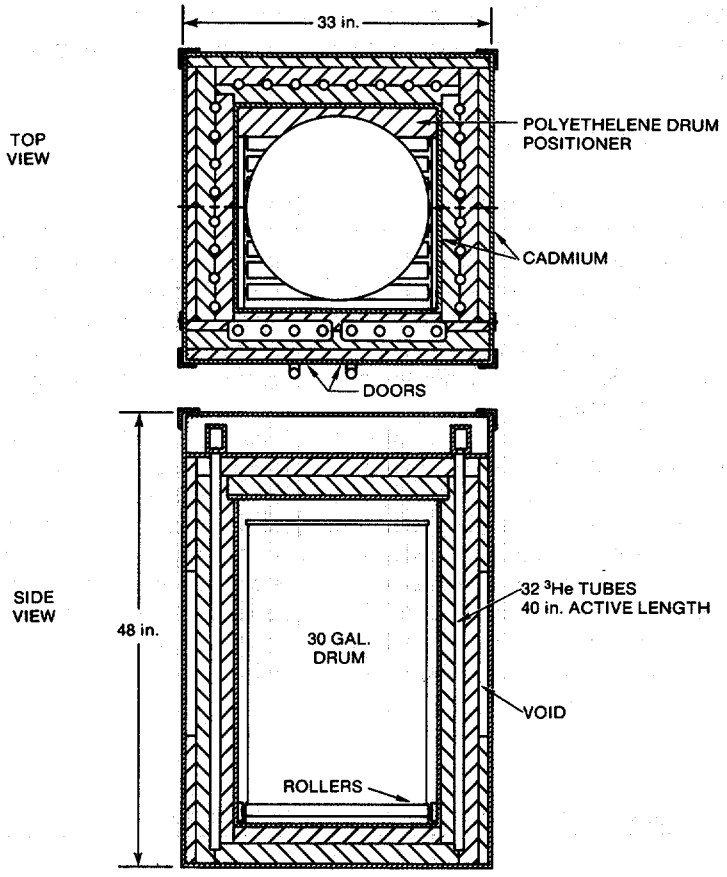

Fig. 20.1 *Cutaway view of one of the Confirmatory Measurement Counters built specifically for shipper and receiver confirmatory measurements of plutonium in 30-gal. shipping drums (Refs. 11 and 12).*

furnaces, gloveboxes, and other equipment can acquire appreciable deposits. When the internal surface areas are large, the total holdup can be enough to affect the plant inventory difference (Ref. 13). The amount of holdup depends on the nature of the process and on the physical form of intermediate solutions, precipitates, and powders. Also, process upsets can lead to large, rapid, and unexpected depositions of material.

Some of the mechanisms for material accumulation (Ref. 14) are summarized below:

(1) gradual sedimentation and settling of fine particles in regions of poor circulation or low flow rate

(2) chemical reaction of nuclear materials with interior walls or migration of the materials into the walls

(3) solid or liquid product formation or precipitation resulting from inadvertent chemical reactions